ARKIV FÖR FYSIK Band 3 nr 13

Communicated 28 February 1951 by LISE MEITNER and ERIK HULTHÉN

On the radioactivity of K^{40}.

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With 10 figures in the text.

1. Introduction

The present investigation was undertaken with the purpose of measuring the branching ratio between electron capture and β-decay in the disintegration of K^{40}. While some of the evidence[1-3] available when these measurements were begun indicated a value of the order of 2 for this branching ratio, determinations of the amount of argon in potassium minerals of known age seemed to yield much lower values, which were of the order of 0.10.[4-7]

As has been pointed out previously[8], the values given by BLEULER and GABRIEL for the branching ratio and for the half-life period of the β-decay would imply an enormous heat production in the past by the radioactivity of the potassium contained in the earth's crust. However, it could be shown subsequently[9], that the value of the β-decay period found by BLEULER and GABRIEL was too low by a factor of about 2 owing to the fact that the influence of back-scattering, which was especially large in their experiment, was not allowed for. A redetermination of the specific β-activity of K^{40} was also made[9], by comparing it to that of UX_2, in an experimental arrangement in which the influence of back-scattering, as well as that of the γ-rays from the two sources, was negligible. The value obtained for the specific activity, 26.8 ± 1.2 β-particles per second and per gram of natural potassium[9], together with the recent value of $(0.0119 \pm 0.0001)\%$[10] for the isotopic abundance of K^{40}, corresponds to a β-decay period of $(1.50 \pm 0.07) \cdot 10^9$ years.[11]

As a consequence of this lower value found for the β-activity, the value of the electron capture to β-process ratio which is given by BLEULER and GABRIEL as 1.9 ± 0.4, should be increased to about 3.8. However, BLEULER

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[1] F. C. THOMPSON and S. ROWLANDS, Nature *152*, 103, 1943.
[2] E. BLEULER and M. GABRIEL, Helv. Phys. Acta *20*, 67, 1947.
[3] L. H. AHRENS and R. D. EVANS, Phys. Rev. *74*, 279, 1948.
[4] P. HARTECK and H. SUESS, Naturwiss. *34*, 214, 1947.
[5] H. E. SUESS, Phys. Rev. *73*, 1209, 1948.
[6] L. T. ALDRICH and A. O. NIER, Phys. Rev. *74*, 876, 1948.
[7] PAHL, HIBY, SMITS, and GENTNER, Z. f. Naturforschung, *5a*, 404, 1950.
[8] E. GLEDITSCH and T. GRÁF, Phys. Rev. *72*, 641, 1947.
[9] T. GRÁF, Phys. Rev. *74*, 831, 1948.
[10] A. O. NIER, Phys. Rev. *77*, 789, 1950.
[11] T. GRÁF, Phys. Rev. *79*, 1014, 1950.

and GABRIEL, in measuring the x-rays from A^{40} (which follow a fraction of the capture processes in K^{40}) employed as selective absorbers different gas layers in which also the absorption of the β-rays differed slightly. In correcting for differences in the β-absorption, BLEULER and GABRIEL used the value of the absorption coefficient corresponding to the primary β-rays of K^{40}. This correction is evidently too small, since it does not allow for the considerable absorption suffered by the soft back-scattered rays. If the absorption of these back-scattered rays is also taken into account, the value for the branching ratio is reduced to about 1.8.[9] In spite of the uncertainty of this correction it seems improbable that it could reduce the value of the branching ratio sufficiently to bring it into agreement with the low values resulting from the argon determinations.

Since the values for the branching ratio obtained by the argon extraction method[4-7] were of the same order of magnitude as the number of γ quanta emitted by K^{40} per β-particle[8], SUESS assumed that the decay scheme of K^{40} is as represented by the full lines of Fig. 1.[5] On this assumption, the capture process would only lead to the excited state of A^{40} and not to its ground state. On the contrary, a large value of the branching ratio[1-3] would imply that also the direct transition to the ground state of A^{40} can occur, as shown by the dashed line of Fig. 1, and that the probability for this transition is even greater than for the transition leading to the excited state.

Recent experiments of BELL and CASSIDY indicate that less than 2.10^{-5} positrons per β-ray are emitted by K^{40}.[12] This result seems to lend additional support to the assumption of SUESS, since it implies that the fraction of the electron capture processes leading to the ground state of A^{40} should be negligible. This follows from the calculations of FIREMAN[13], which, for a K^{40}–A^{40} mass difference corresponding to 1.65 MeV, yield a value of about 160 for the probability ratio between electron capture and positron emission. The energy difference between the ground states of K^{40} and A^{40} is obtained by adding 0.30 ± 0.08 MeV, corresponding to the Ca^{40}–A^{40} mass difference as measured by ROBERTS and NIER[14], to the maximum energy of the β-rays, 1.35 ± 0.03 MeV.[15-17] For a γ-ray energy of 1.46 ± 0.02 MeV[12, 18] the excited state of A^{40} appears thus to lie 0.19 ± 0.09 MeV below the ground state of K^{40}.

In a previous note[11], a brief description was given of the experimental method used in the present investigation, together with some preliminary results indicating an upper limit of about 0.7 for the branching ratio. Although this value is definitely smaller than those of references 1 to 3, it still differs from the γ/β ratio more than would seem to be compatible with the experimental errors.

In order to clear up this discrepancy, the main measurements have been repeated several times. They have been completed by a number of control experiments, for the purpose of eliminating possible secondary effects. These

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[12] P. R. BELL and J. M. CASSIDY, Phys. Rev. *79*, 173, 1950.

[13] E. L. FIREMAN, Phys. Rev. *75*, 1447, 1949.

[14] T. R. ROBERTS and A. O. NIER, Phys. Rev. *79*, 198, 1950.

[15] DZELEPOW, KOPJOVA, and VOROBJOV, Phys. Rev. *69*, 538, 1946.

[16] BELL, WEAVER, and CASSIDY, Phys. Rev. *77*, 399, 1950.

[17] D. E. ALBURGER, Phys. Rev. *78*, 629, 1950.

[18] PRINGLE, STANDIL, and ROULSTON, Phys. Rev. *77*, 841, 1950.

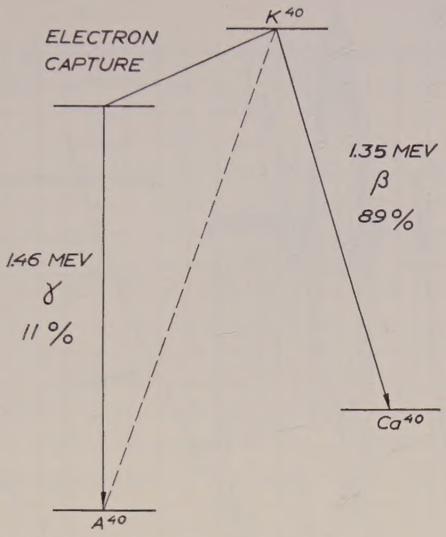

Fig. 1. Decay scheme of K^{40}.

measurements are described in section 4, while the corrected values obtained for the branching ratio are discussed, together with some recent values published by other experimenters, in section 6, at the end of this paper.

Section 2 contains a description of the experimental method used, as well as some remarks on the use of G—M counters in measurements of high accuracy. Section 3 deals with the calculation of the x-ray efficiency for an internal-source G—M counter of the type used in this work, and gives a simple approximate formula for this efficiency. In section 5, the β-efficiency of the counter and the corrections necessitated by the variations of this efficiency are considered.

2. Experimental method

2 a. Principle of the method

The number of K-capture processes per second in K^{40} may be determined by detecting either the x-rays or the Auger electrons which follow the capture process. For argon the K fluorescence yield is only about 12 %, the probability of the Auger process being thus 88 %. The range of the Auger electrons is, however, less than 0.1 mg/cm^2, and the intensity from a source of natural potassium of this thickness would be too weak to be measurable. Consequently, an x-ray method, similar to that of BLEULER and GABRIEL[2], was employed, but in which instead of changing critical absorbers placed outside the counter, the filling gas of the G—M counter itself was used as selective detector.

The source was placed directly on the inside walls of the counter which was filled alternately with two gases, one possessing low, the other high efficiency for the K_α x-rays of argon. The mass absorption coefficient of these rays,

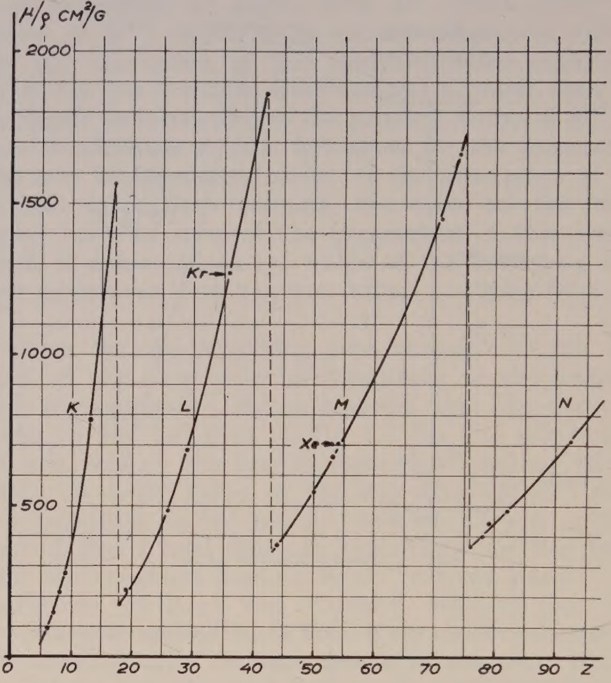

Fig. 2. Mass absorption coefficient of the K_α x-rays of argon (4180 X.U.) in the different elements.

computed with the help of the tables of Jönsson[19], is shown in Fig. 2 as a function of the atomic number of the absorber. As is seen, the most suitable high-efficiency gas is krypton, which has a larger mass absorption coefficient than xenon, and also a larger linear absorption coefficient, in spite of its smaller density. As low-efficiency gas, argon was chosen.

Both argon and krypton make excellent filling gases for G–M counters. They both possess high primary specific ionization for β-rays. It seemed, therefore, reasonable to assume that, at the same pressure, the β-efficiency of the counter for the two gases would be the same within 0.1 %. Since the γ radiation of K^{40} is very little absorbed in the filling gases, its contribution to the counting rates can easily be shown to be negligible. It is certainly less than 0.01 % of the total rate for the thin sources used in this work. In these conditions, the counting rate differences deduced from alternate measurements made with the two gases should give a measure of the x-ray intensity.

Computation of the number of x-ray counts/min., carried out in the way outlined in section 3, yielded a rate difference of about 2 %, when assuming a branching ratio equal to one. Since previous experience with G–M counters has shown that, with sufficient care, single measurements at low counting rates may be made with an accuracy of about 0.1 %, the determination of a branching ratio of the order of two appeared to be a relatively easy task. It was clear,

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[19] E. Jönsson, Inaugural dissertation, Uppsala, 1928.

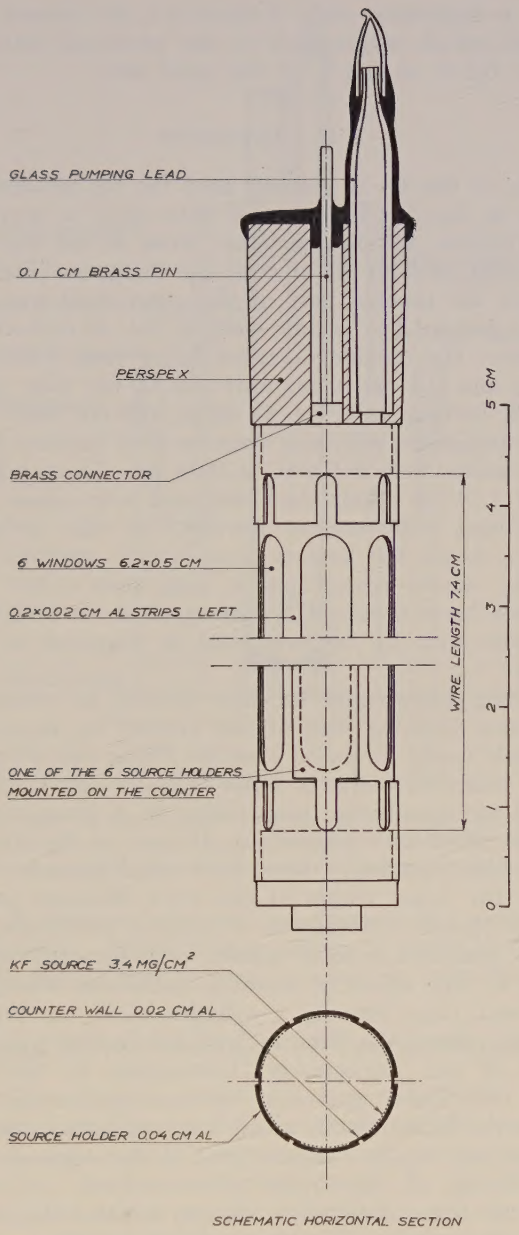

Fig. 3. Schematic view of the GEIGER-MÜLLER counter showing arrangement of the six source holders.

however, that for a branching ratio of about 0.1, the present method could only give an upper limit which corresponds to the combined statistical error of two measurements, i.e., 0.2 % to 0.3 % of the total rate.

2 b. Apparatus

The construction of the G–M counter used for the measurement of the x-ray intensity is shown in Fig. 3. The counter tube itself is supported by a tapered plug of Perspex (shown in cross-sectional view at the top of Fig. 3), and is placed inside an outer shell of Al of 0.02 cm thickness (Fig. 4). The joint between the plug and the tapered part of the outer shell was made gas-tight by means of stopcock grease. The middle part of the Al cathode was turned down to 0.02 cm thickness, the outside diameter being then 1.34 cm. Six large windows, 6.2 cm long and 0.5 cm wide, were cut in the thin part of the cathode, as shown in Fig. 3, leaving only six Al strips, 0.2 cm wide. The source, in the form of a natural potassium salt, was evaporated in vacuum onto six Al holders, the dimensions of which were 6.4 × 0.7 × 0.04 cm. These holders were cut from an Al cylinder, of 1.34 cm inside diameter, and were made with a tab at each end. The tabs fitted into recesses provided in the cathode cylinder, and made it possible to locate the holders reproducibly over the cathode windows. In this design the source constitutes a large part of the inside wall of the counter. The source layer occupied a surface of about 5.92×0.49 cm^2 on each holder. The counter wire of tungsten had a diameter of 0.01 cm. Its free length was 7.4 cm.

Changing the filling gases could be done quickly by changing the small glass tube attached to the pumping lead of the counter by means of Apiezon wax. This glass tube could easily be sealed once the filling was completed. It occupied less space than a stopcock, and was easier to make light-tight. The counter was pumped for 10–20 minutes before each filling to a pressure of about 0.001 mm of Hg. It was first filled with alcohol, at 15 mm of Hg, then with one of the rare gases, which were chemically pure, to a total pressure of 200 mm of Hg, at 20° C. During the later stages of the work different pressures were used.

The counter was placed vertically in a support which could also hold one of several cylindrical standard sources, placed concentrically around the counter, as shown in Fig. 4. The effect of external radiations was greatly reduced by means of large lead rings forming a cylinder of 10 cm thickness around the counter. Under and above the counter lead bricks were placed to a total thickness of 7.8 and 10 cm, respectively. Connexions to the amplifier were led through a small rectangular gap left between two of the upper lead bricks. In order to avoid the light effects which were observed during the first runs, both this gap and the upper Perspex plug of the counter were covered with opaque materials during all subsequent measurements.

The impulses from the counter were fed into a Neher-Harper circuit connected to a conventional scale-of-64. The high voltage for the counter was furnished by a battery-biased stabilizer. This voltage could be kept constant to within 1 or 2 volts throughout the whole day. The starting voltage of the counter could be determined with the same precision. A constant overvoltage of 60 volts was furnished by a dry battery connected in series with the high voltage stabilizer.

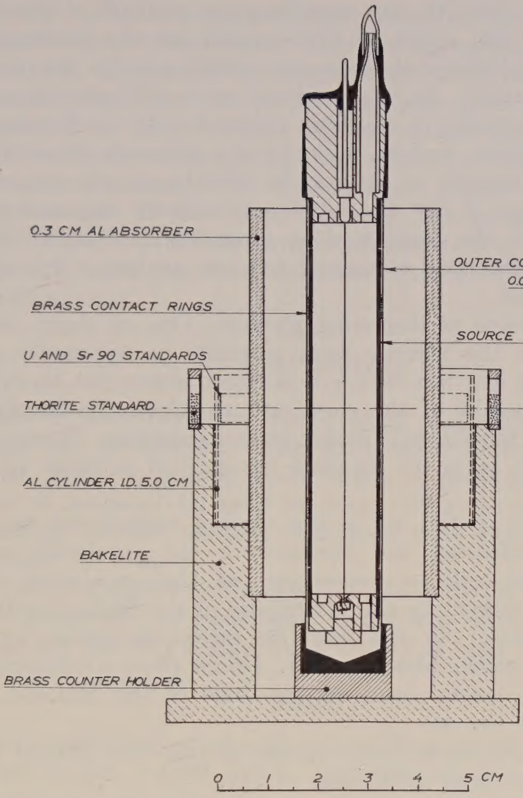

Fig. 4. Cross-sectional view of the counter with the concentrically arranged standard sources. The Al absorber cylinder is only used together with the thorite standard. For convenience both types of standard sources are represented on the same diagram.

Two telephone call counters were used simultaneously in order to get a constant check on their operation. Without this artifice it would be difficult to discover in time a slight slackening due to some mechanical imperfection.

2 c. Remarks on the operation of GEIGER-MÜLLER counters

In order to obtain sufficient statistical accuracy with the low counting rates yielded by the potassium sources, long counting periods were necessary. This fact offered a good opportunity for studying and improving the reliability of the counter operation. In fact, the method used in the present investigation depends to a very great extent on this reliability, since the alternate measurements had to be reproducible to within about 0.1 %, even after repeated changes of the filling gases. Some of the methods used for improving and checking the counter operation will be given here.

It was found convenient to continue the measurements 24 hours a day, and to leave the scale and the high voltage apparatus on the mains for periods of several months at the time. This practice not only reduced the variations in room temperature due to the switching on and off of the electric power, but it also eliminated the waste of time caused by the heating periods during which the operation of the counter is not stable enough for measurements of great precision to be made. It is considered to be still more important to avoid all effects which may result in spurious pulses in the G—M counter.

One cause of spurious counts may be the presence of air in the counter. Comparatively large amounts of air may be introduced into the counter together with the alcohol vapour if the alcohol is not carefully degassed prior to its use. Thus, the increase of the plateau slope observed by SPATZ[20] with increasing concentrations of alcohol was explained by him as being probably due to air dissolved in the alcohol.

There are several ways of degassing alcohol. One of them, which has been used by the author in the present work, consists in repeatedly pumping away the air-alcohol vapour mixture which is formed above the liquid alcohol after each pumping. On account of the slow rate at which the dissolved air diffuses out of the liquid this procedure is rather time consuming. It may be interrupted too early if no care is taken to measure the partial pressure of the air in the alcohol vapour.

The partial pressure of the air in the alcohol vapour can be determined in a simple way by measuring the pressure of the air-alcohol mixture with a McLeod manometer or with a vacuscope, at the saturation pressure of the alcohol. Part of the vapour in the closed end of the McLeod will condense, but the vapour pressure there will still have its saturation value, i.e., the same as on the side connected to the filling system. Under these conditions the difference between the mercury levels, read on the scale of the McLeod, gives directly the partial pressure of the air.

It was checked during each filling operation that the partial pressure of air in the alcohol was less than 0.005 mm of Hg.

Another possible cause of spurious pulses is the presence of water vapour in the counter. The observation, that with a dry KF source inside the counter a plateau slope of about 2 % per 100 volts could be maintained for several weeks, suggests the possibility that the use of a drying agent could improve the counter characteristic, at least when as here, the counter has not been heated while on the vacuum system.

It is, of course, necessary to check the absence of spurious pulses even when all precautions have been taken during the filling of the counter. This checking was done with the anticoincidence amplifier* which was used to reduce the background counts of the previously described multiple γ-counter.[21] The sensitivity of the anticoincidence channel of this amplifier was lower than that of the direct channel, and it was found that, when the anode of the counter was connected simultaneously to both channels, a small fraction of the single counting rate, of the order of 0.1 %, was still registered. The fraction counted increased in some cases with prolonged use of the counter from about 0.1 % to several percent. It could be shown that this increase was due to the increase in the number of spurious pulses of the counter, though only a fraction of the spurious pulses was registered in this way.

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[20] W. D. B. SPATZ, Phys. Rev. *64*, 236, 1943.

* The author is indebted to Mr. G. VON DARDEL for the loan of the anticoincidence amplifier.

[21] T. GRÁF, Rev. Sci. Inst. *21*, 285, 1950.

This behaviour of the anticoincidence amplifier may be attributed to the fact that pulses which follow closely a preceding pulse are smaller than normal, and spurious pulses may therefore trigger only the direct channel of the amplifier. Other causes than spurious pulses may also give a small number of anticoincidence counts, but their number will not change with the use of the counter. More elaborate methods of measuring the number of spurious counts have been published by several authors.[22-24] The standard source used for this check gave about 800 counts/min. in the direct channel. The measurements with K^{40} were only accepted if the number of anticoincidence counts at the start of the run was less than 0.1 % of this total rate, and showed no increase at the end of the main measurements.

2 d. Sources.

The K^{40} sources were made by evaporating chemically pure potassium salts in vacuum onto the Al holders.* The six holders were placed side by side on a curved brass plate, in a way which made it possible to deposit practically equal amounts of material on each of them. Only the central part of the holders, corresponding to the cathode windows, were exposed, their edges being protected by thin Al foils.

One of the most suitable substances for the purpose in view is KF. It contains a large proportion of potassium and has a relatively low mass absorption coefficient for the K_α x-rays from argon, as may be seen from Table 1.

Table 1

Mass absorption coefficient of the K_α x-rays from argon in different substances. Wave length = 4180 X. U. Quantum energy = 2.96 keV.

Substance	A	Kr	KF	KCl	C	Al
μ/ϱ cm²/g	174	1270	240	860	96	790

Since it is very hygroscopic, however, precautions have to be taken to keep it dry. Before mounting, the KF sources were kept in a desiccator. During the weighings, some $CaCl_2$ was placed on a grid, immediately above the sources, and during mounting the counter and the source holders were kept in contact with a large metal plate, the temperature of which was held at about 60°C.

According to a rough preliminary calculation, the counting time necessary to detect a given x-ray intensity with the present experimental arrangement, has a minimum value for a source thickness of 3 to 4 mg/cm². Therefore, in the main measurements with KF, a source of 3.44 mg/cm² was used, containing a total amount of 40.5 mg of K. After weighing, the source layers were painted with colloidal graphite in alcohol suspension. This was done in order

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[22] S. C. Curran and E. R. Rae, Rev. Sci. Inst. *18*, 871, 1947.

[23] J. L. Putman, Proc. Phys. Soc. London *61*, 312, 1948.

[24] D. Willard and C. G. Montgomery, Rev. Sci. Inst. *21*, 520, 1950.

* The author is indebted to Mr. J. E. Flinta for carrying out the evaporations.

to improve the operation of the counter, and to avoid the spurious counts which may be due to electron emission from electrically charged thin insulator layers.[25] The average thickness of the graphite layer was about 0.3 mg/cm^2, the corresponding x-ray absorption being of the order of a few percent.

Although the counter operation was very good at the start, after several changes of the filling gas spurious counts appeared. Since an examination of the sources revealed the presence of small white spots on their surfaces, the sources were painted over with an additional thin graphite layer. When the number of spurious counts increased again progressively after successive fillings of the counter, it became apparent that the white spots were caused by the filling process itself. It seemed likely that flushing the counter with saturated alcohol vapour, as was practised before each filling, might move some of the weakly bound colloidal graphite particles from their original place on the surface of the source. This practice was therefore abandoned, with the result that no spurious effects were observed after the subsequent fillings.

After the main runs with KF it appeared desirable to study the variation of the β-counting rate alone, as a function of the rare gas pressure. For this purpose KCl was used, in which substance the K_α x-rays of argon are strongly absorbed. In order to secure a β-counting rate practically equal to that given by the KF source, the potassium content of the KCl layer had to be the same as that of the KF source. The KCl source used had a thickness of 4.12 mg/cm^2, and contained 37.9 mg of K. It was deposited on six Al holders similar to those which were used for the KF source. A trial experiment made with an uncovered KCl source gave no spurious counts, showing that the use of graphite was not necessary. The KCl measurements were therefore made without graphite on the source.

The counter operation was checked at the end of each run by means of standard sources. The active substance in these sources was evenly distributed on cylindrical holders. Each standard source was placed concentrically around the counter, and symmetrically with respect to its median plane (Fig. 4). With this arrangement the counting rates were reproducible to within an accuracy of about 0.1 %.

The following substances were used:

1. **Uranium oxide.** About 105 mg of finely ground UO_2 was deposited evenly on the inside wall of an Al holder of 5.0 cm diameter, in the form of a cylindrical band of 0.8 cm width. The source was fixed to the holder by means of cellulose acetate, and was covered by a thin layer of the same substance. The radiation entering the counter consists almost exclusively of the UX_2 β-rays, of maximum energy 2.32 MeV. A small part of it consists of the L fluorescence radiation (740—770 X. U), emitted in about 5 % of the UX_1 desintegrations.[26] This radiation is absorbed to different extents in krypton and argon, and may therefore give slightly different counting rates with the two gases.
2. **Sr^{90}.** A diluted aqueous solution of $SrCl_2$, containing Sr^{90},* was deposited evenly and dried on the inside wall of a holder similar to the one used for UO_2. The width of the surface covered was 0.6 cm. The radiation entering the

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[25] H. Paetow, Z. f. Phys. *111*, 770, 1939.

[26] H. Bradt and P. Scherrer, Helv. Phys. Acta *19*, 307, 1946.

* The $SrCl_2$ solution was kindly provided by Mr. T. Vestermark.

counter consists mainly of the 2.3 MeV β-rays from Y^{90}, a daughter substance of the long-lived Sr^{90} which emits β-rays of only 0.6 MeV maximum energy. No γ radiation is emitted by these substances. A recent determination gives 19.9 ± 0.3 years for the half-life period of Sr^{90}.[27]

3. Thorite. This source, consisting of 4.8 g of the mineral placed in a ring-shaped container, was to be used as a γ-ray standard for checking the counter operation when the same fillings but different internal absorbers were employed. For this reason, the thorite standard was always used conjointly with an external Al absorber of 0.3 cm thickness. Both the standard and the absorber are indicated in Fig. 4.

Before describing the measurements made with the K^{40} sources, the method of calculation will be given which was used to compute the x-ray counting rates to be expected for the different experimental conditions encountered.

3. Computation of the counting efficiency for x-rays

3 a. General considerations on the counting efficiency of Geiger—Müller counters for soft x-rays

The absorption of x-rays of quantum energy of the order of a few keV is almost exclusively due to the photoelectric process. In this process, the whole energy of a quantum is expended in extracting an electron from one of the extra-nuclear shells of an atom, and in communicating to the electron its kinetic energy.

Scattering of x-rays, by Compton effect, is comparatively rare in the low energy region. It is at least 1000 times less frequent than absorption by the photoelectric effect. For this reason, scattering may be neglected, and the path of a soft x-ray regarded as a straight line ending at the point where the quantum is absorbed.

When the act of absorption takes place in the filling gas of a counter, the photoelectron extracted from the K or L shell of a gas atom will give rise to a great number of secondary electrons. Thus, the ionization produced in the filling gas by the absorption of a quantum of low energy may always be regarded as amply sufficient for initiating a count.

The length of the effective counting volume, in which, by definition, the presence of a single slow electron is sufficient to produce a count, is not known exactly. An upper limit for its extension in the axial direction is the distance between the plane faces of the end plugs. In the present construction the distance between these faces is equal to the free wire length, as shown in Fig. 4. As a lower limit, it may be assumed that the counter volume is bounded by two transversal planes situated at a distance equal to one-tenth of the free wire length from the end plugs. This distance corresponds to 1.1 times the counter radius.

Experimental investigations of H. Saurer[28] on the efficiency of G—M counters for soft x-rays show, that with a wire support similar to the one used here (diameter 2 mm), the efficiency along the counter axis begins to decrease at a distance corresponding to about 0.8 of the counter radius from the supports, and that its decrease is 50 % at approximately half this distance.

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[27] R. I. Powers and A. F. Voigt, Phys. Rev. *79*, 175, 1950.

[28] H. Saurer, Helv. Phys. Acta *23*, 381, 1950.

The quantum efficiencies, computed with the two limiting assumptions made concerning the length of the effective counter volume, were found to have values differing by about 5 % from each other. It seems reasonable to assume that the mean of the two values corresponds to the true effective volume of the counter to within less than 2 %.

Owing to the exceedingly short range in solid matter of the photoelectrons produced in the counter walls (0.02 to 0.05 mg/cm²), their contribution to the total quantum efficiency is small. Since, in addition, this wall effect is independent of the nature of the filling gas, it does not influence the final results of measurements in which readings taken with different gases enter only by their difference.

3 b. Computation of the number of x-ray quanta counted in the counter

Owing mainly to the absence of noticeable scattering in the case of low-energy quanta, the number of counts produced by the x-rays from a thin cylindrical source layer can readily be computed with sufficient accuracy. Since the need for similar computations may arise also in investigations concerning other cases of electron capture, it is thought worth while to give here some details on the derivation of the necessary formulae.

It will be assumed in the following calculations that absorption within the thin source layer, and in a thin absorber foil in contact with it, may be represented by the analytical expression corresponding to the case of a flat source. This assumption was verified by computing the error due to the curvature of the source layer under the conditions used here. The error was found to be entirely negligible.

Let n_0 be the number of quanta emitted per gram of source material per second, ϱ the density, and t the thickness of the source layer, and imagine a solid cylinder of unit cross section, and of length equal to t, laid off inside the source, as shown in Fig. 5. A cylindrical volume element of thickness dx, lying at a distance x from the surface of the source, emits $n_0 \varrho \, dx$ quanta per second. The number of quanta entering the counter volume from the cylinder within a solid angle $d\Omega$, in the main direction θ relative to the normal to the surface of the source, is

$$d n_\theta = \frac{n_0 \cdot \varrho \cdot d\Omega}{4\pi} \cdot \int_0^t p \cdot dx . \tag{1}$$

In this expression p is the probability that a quantum will *not* be absorbed in the source, or in the absorber.

The integral in expression (1) can be solved at once. The probability p is

$$p = e^{-(\mu_s \cdot x + \mu_a \cdot a)/\cos\theta} , \tag{2}$$

with a = thickness of the absorber, and
μ_s, μ_a = linear absorption coefficients of the radiation in the source and the absorber, respectively.

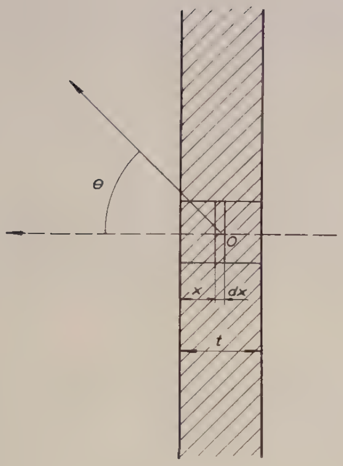

Fig. 5. Schematic view representing the cross-section of the source layer.

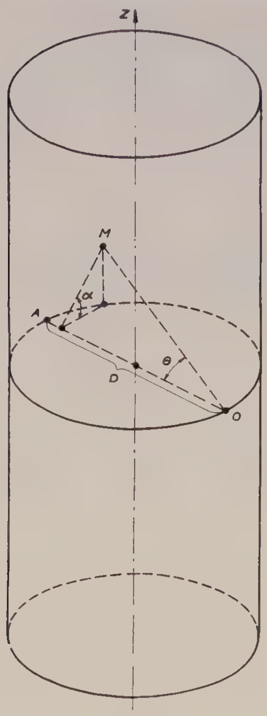

Fig. 6. Diagram representing the counter volume and the path OM of an x-ray quantum in the counter.

The result of the integration is

$$d\,n_\theta = \frac{n_0 \cdot d\,\Omega}{4\,\pi \cdot (\mu_s/\varrho)} \cdot (1 - e^{-\mu_s \cdot t/\cos\theta}) \cdot e^{-\mu_a \cdot a/\cos\theta} \cdot \cos\theta\,. \tag{3}$$

The solid angle $d\,\Omega$ entering into expression (3) may be written in the form

$$d\,\Omega = \sin\theta \cdot d\,\theta \cdot d\,a \tag{4}$$

in which the angle a, shown in Fig. 6, is the dihedral angle between a plane perpendicular to the axis of the counter and the plane containing the path of the ray, OM, and the normal at O.

The counting efficiency, i.e., the probability of a quantum being absorbed in the counter gas, depends on the length, l_θ, of its path inside the effective counting volume, and on the linear absorption coefficient, μ, of the radiation in the filling gas at the pressure used. It is given by the expression

$$\varepsilon_\theta = 1 - e^{-\mu \cdot l_\theta}. \tag{5}$$

The path length itself is a function of θ, α, and the axial coordinate z of the point of emission of the quantum.

The number of counts, N_x, due to the x-rays from the whole surface, $S = L \cdot b$, of the source, may be obtained by multiplying the number of quanta from the surface element $b \cdot dz$ emitted within the solid angle $d\Omega$,

$$d n_\theta \cdot (S/L) \cdot dz,$$

by the efficiency ε_θ, and by integrating with respect to the variables α, z, and $\cos\theta$, which latter is more convenient to use than θ.

The expression giving N_x is

$$N_x = \frac{n_0 \cdot S}{2(\mu_s/\varrho)} \cdot \int_{\cos\theta=0}^{\cos\theta=1} (1 - e^{-\mu_s \cdot t/\cos\theta}) \cdot e^{-\mu_a \cdot a/\cos\theta} \cdot \cos\theta \cdot \left[\frac{1}{L} \cdot \int_0^L \frac{1}{2\pi} \cdot \int_0^{2\pi} (1 - e^{-\mu \cdot l_\theta}) \cdot d\alpha \cdot dz\right] \cdot d(\cos\theta). \qquad (6)$$

Written in this form, this expression allows one to recognize immediately the factor within the square brackets as the mean value of the counter efficiency, $\bar{\varepsilon}(\cos\theta)$, averaged for a given value of $\cos\theta$, over all possible values of α and z.

In order to calculate $\bar{\varepsilon}(\cos\theta)$, an explicit expression of l_θ is required, in terms of θ, α and z. Such an expression may easily be obtained, for l_θ is simply the distance OM between two points on the cylindrical surface, or the distance of a point on this surface to a point on one of the end planes.

In the first case, i.e., when the point M is on the cylindrical surface, of diameter D, the value of l_θ is

$$l'_\theta = D/(\cos\theta + \sin\theta \cdot \mathrm{tg}\,\theta \cdot \cos^2\alpha). \qquad (7)$$

In the second case, when M is on one of the bounding planes, situated at a distance z from O, it is given by

$$l''_\theta = z/(\sin\theta \cdot \sin\alpha). \qquad (8)$$

When for a given value of $\cos\theta$, the rays originating from the point O may reach one of the bounding planes, the region of integration with respect to α must be subdivided into 4 subregions, in which l_θ is given either by (7) or by (8), and the limits of which depend on the distance of the bounding plane from the point O.

Thus, when both bounding planes may be reached by the rays, the expression of $\bar{\varepsilon}(\cos\theta)$ takes the form

$$\bar{\varepsilon}(\cos\theta) = \frac{1}{L}\int\limits_0^L \frac{1}{\pi}\Bigg[\int\limits_0^{\alpha_1}(1 - e^{-\mu\cdot l'_\theta})\,d\alpha + \int\limits_{\alpha_1}^{\pi/2}(1 - e^{-\mu\cdot l''_\theta})\,d\alpha +$$

$$+ \int\limits_0^{\alpha_2}(1 - e^{-\mu\cdot l'_\theta})\,d\alpha + \int\limits_{\alpha_2}^{\pi/2}(1 - e^{-\mu\cdot l''_\theta})\,d\alpha\Bigg]\cdot dz. \qquad (9)$$

The limits α_1 and α_2 correspond to the case of a path reaching the circle of intersection between the cylindrical surface and either bounding plane. They are defined by the condition $l'_\theta = l''_\theta$, yielding the expression

$$\alpha_i = \arc \sin\left\{\frac{D}{2\,z_i\cdot \operatorname{tg}\theta}\left[\sqrt{1 + \left(\frac{2\,z_i}{D\cos\theta}\right)^2} - 1\right]\right\}, \qquad (i = 1, 2) \qquad (10)$$

in which z_i denotes the distance of either bounding plane from the point O.

Expression (6), giving the counting rate due to x-rays from the whole source, may be written in the following compact form:

$$N_x = \frac{n_0\,S\,I_0}{2\,(\mu_s/\varrho)}, \qquad (11)$$

with

$$I_0 = \int\limits_0^1 f_s\cdot f_a\cdot\bar{\varepsilon}(\cos\theta)\cdot\cos\theta\cdot d(\cos\theta), \qquad (12)$$

where

$$f_s = (1 - e^{-\mu_s\cdot t/\cos\theta}), \quad \text{and} \quad f_a = e^{-\mu_a\cdot a/\cos\theta}.$$

Thus, the problem is reduced to the computation of the simple integral I_0. This computation may be carried out to any desired degree of accuracy by numerical integration.

The computation may be considerably simplified by observing that the average value of the counter efficiency, ε_{av}, which is given by

$$\varepsilon_{av} = I_0 \Big/ \int\limits_0^1 f_s\cdot f_a\cdot\cos\theta\cdot d(\cos\theta), \qquad (13)$$

may be represented approximately by the simple expression

$$\varepsilon_{av} \approx \text{const}\cdot(1 - e^{-\mu\cdot D}). \qquad (14)$$

It was verified by computing expression (13) for the different sources and rare gas pressures employed in this work, that the value of the constant in (14) is 0.91, with an uncertainty of about 1 %. The simple formula (14) giving the average x-ray efficiency of a G—M counter for an internal source may be used in similar cases if no great accuracy is required, though the value of the constant will be different from 0.91.

With the approximate value of ε_{av} given by (14), the computation of the integral

$$I_0 = \varepsilon_{av} \cdot \int_0^1 f_s \cdot f_a \cdot \cos\theta \cdot d(\cos\theta) \tag{15}$$

becomes an easy matter.

The counting rates due to the K_α x-rays from argon were computed with the help of formulae (11) and (15) for the two different sources and the various pressures used in the present work. The results of the computations are summarized in Tables 2 a and 2 b.

Table 2 a

Computed number of counts/min. due to the K_a x-rays from argon.
KF source of 3.44 mg/cm². S = 17.58 cm².

Absorber	Rare gas pressure at 20° C in mm of Hg	μ cm^{-1}	Average x-ray efficiency in %	Integral I_0	$N_x = \frac{n_0 S I_0}{(2\mu_s\varrho)}$ Counts min. for a branching ratio of	
					1	0.127
0.7 mg/cm² graphite . .	Kr 128	0.739	0.569	0.180	0.78	0.100
» » . . .	Kr 185	1.069	0.689	0.217	0.95	0.121
» » . . .	A 185	0.070	0.083	0.026	0.11	0.015
0.6 mg cm² graphite +2.3 mg/cm² Al	Kr 185	1.069	0.689	0.017	0.08	0.01
»	A 185	0.070	0.083	0.002	0.01	0.001

Table 2 b

Computed number of counts/min. due to the K_a x-rays from argon.
KCl source of 4.12 mg/cm². S = 17.55 cm².

Absorber	Rare gas pressure at 20° C in mm of Hg	μ cm^{-1}	Average x-ray efficiency in %	Integral I_0	$N_x = \frac{n_0 SI_0}{2(\mu_s\varrho)}$ Counts/min. for a branching ratio of	
					1	0.127
0	Kr 128	0.739	0.569	0.284	0.27	0.034
0	Kr 185	1.069	0.689	0.344	0.33	0.041
0	Kr 388	2.24	0.86	0.43	0.41	0.052
0	A 185	0.070	0.083	0.042	0.04	0.005
0	A 564	0.214	0.23	0.115	0.11	0.014
1.0 mg/cm² Al	Kr 185	1.069	0.689	0.10	0.09	0.011
»	Kr 388	2.24	0.86	0.13	0.12	0.015
»	A 564	0.214	0.23	0.04	0.04	0.005

The presence of the K_β x-rays from argon (3880 X. U.), which are less absorbed in the counter gases than the K_α rays, has no appreciable influence on the results, for the lower efficiency of the counter for these rays is nearly compensated by the fact that the absorption of the K_β rays in the source layer is also smaller. Moreover, their intensity represents only about 12 % of the total intensity due to the K series x-rays.

For the K fluorescence yield in argon the value of 0.12 was taken. It represents an average of the experimental values quoted by COMPTON and ALLISON.[29] More recent experiments indicate the appreciably lower values of 0.077[30], and 0.06.[31] There is thus a rather large uncertainty in the value of the fluorescence yield. However, the results deduced for the branching ratio may easily be corrected for any value which would appear to be more reliable than that adopted here.

In order to obtain the total probability of electron capture in K^{40}, the capture of an electron from the L_I shell must also be considered. For the L_I/K capture probability ratio computations of ROSE and JACKSON give a value of 8.2 %.[32] The contribution of the other shells is negligible.

For the specific β-activity of potassium the previously determined value of 26.8 ± 1.2 β-rays per gram of potassium per second was used.[9] Since this determination was made, the specific β-activity of potassium has been measured by a number of experimenters who obtained values varying between 25 and 32 β/g/second, with a weighted mean value of about 28 β/g/second. The results of the most recent determinations, 28.3 ± 1.0 β/g/second[33], and 27.1 ± 1.5 β/g/second[34], also agree with the adopted value, within the limits of the experimentals errors.

4. Measurements

4 a. Measurements with KF

The background rate of the counter with both krypton and argon fillings was determined before evaporating the KF layers onto the six Al holders. These

Table 3

Background measurements made without scaling circuit

Rare gas pressure in mm of Hg	Number of counts	Counting time in min.	Counts/min.
A 185	26 642	3 160	8.43 ± 0.05
Kr 185	45 192	5 359	8.43 ± 0.04
		Difference	0.00 ± 0.07

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[29] A. H. COMPTON and S. K. ALLISON, *X-rays in Theory and Experiment* (D. van Nostrand Company, Inc., New York, 1935), second edition, p. 489.

[30] MARTIN, BOWER, and LABY, Proc. Roy. Soc. A *148*, 40, 1935.

[31] G. M. INSCH, Phil. Mag. *41*, 857, 1950.

[32] M. E. ROSE and J. L. JACKSON, Phys. Rev. *76*, 1540, 1949.

[33] G. A. SAWYER and M. L. WIEDENBECK, Phys. Rev. *79*, 490, 1950.

[34] HOUTERMANS, HAXEL and HEINTZE, Z. f. Phys. *128*, 657, 1950.

measurements gave in both cases 8.43 counts/min., with a combined standard deviation of 0.07 counts/min. The results of these measurements are indicated in Table 3, together with the counting times and the total number of impulses counted in each case.

In order to check the identity of the β counting rates with the two fillings, measurements were made with the KF source described in subsection 2 d, an Al foil of 2.3 mg/cm^2 being placed between the source holders and the cathode windows of the counter. According to the data listed in Table 2 a, this foil absorbs almost completely the argon K$_\alpha$ x-rays emitted by the KF source in the direction of the counting volume. The results of these measurements are given in rows Nos. 1 and 2 of Table 4. As is seen, the results obtained with the two gases were identical within a combined standard deviation of 0.11 counts/min. The starting voltage of the counter was about 1000 volts with argon and 1200 volts with krypton.

Table 4

Measurements made with a KF source of 3.44 mg/cm^2

No.	Absorber	Rare gas pressure at 20° C in mm of Hg	Total counting time in min.	Total number counted	Counts/min.	Diff. Kr–A counts/min.
1	0.6 mg/cm^2 graphite + 2.3 mg/cm^2 Al	A 185	8 904	399 495	44.87 ± 0.07[1]	—
2	»	Kr 185	5 040	226 133	44.87 ± 0.09	0.00 ± 0.11[2]
3	0.7 mg/cm^2 graphite	A 185	22 552	1 053 421	46.71 ± 0.05	—
4	»	Kr 185	23 592	1 111 887	47.13 ± 0.04	0.42 ± 0.06
5	»	Kr 128	14 336	670 655	46.78 ± 0.06	0.07 ± 0.08

[1] This value may be too high by as much as 1 % owing to a possible light-effect.
[2] For the reason given under [1]) this value may be as high as 0.4 counts/min.

The standard sources, measured at the end of each KF run, gave the counting rates listed in Table 5. The different runs are referred to by the corresponding Nos. of Table 4. The counting rate yielded by the Sr^{90} standard at

Table 5

Measurements made with standards at the ends of the corresponding runs of Table 4

No.	Rare gas pressure at 20° C in mm of Hg	Internal Al absorber mg/cm^2	Thorite 3 mm Al absorber γ counts/min.	UO$_2$ No absorber β counts/min.	Sr^{90} No absorber β counts/min.
1	A 185	2.3	745 ± 3	1935 ± 4	> 5230
2	Kr 185	2.3	772 ± 3	1932 ± 8	5216 ± 13
3	A 185	0	738 ± 2	1896 ± 4	5164 ± 7
4	Kr 185	0	773 ± 2	1916 ± 4	5153 ± 6
5	Kr 128	0	757 ± 2	1898 ± 6	5154 ± 9

the end of run No. 1 was found to increase from its initial value of 5230 counts/min. to about 5270 counts/min. after 48 minutes of counting. This variation was at first attributed to an increase of the background of the counter caused by the relatively high counting rate. Such an effect has previously been found by LAUTERJUNG and NEUERT[35] and by CHAUDHRI and FENTON.[36] Since the same authors found that fast counting also increased the photosensitivity of the counter, the upper Perspex plug of the counter was, after this run, covered with more wax, and the gap in the lead shielding was also made light-tight. However, no systematic error was suspected in the corresponding result obtained with KF (Table 4, row No. 1), because the counting rate in this measurement was low and, besides, was in excellent agreement with the result of the krypton measurement given in row No. 2. Nevertheless, when examining the data at the end of all the KF measurements, it became apparent that external light must have increased the KF counting rate during run No. 1 also, possibly by as much as 1 %. This is suggested also by the results obtained with the other standards. Thus, the counting rate for the thorite standard was about 1 % higher in run No. 1 than in run No. 3, although the counter gas was argon in both of these runs, whereas the results Nos. 2 and 4, obtained with krypton, were in good agreement. The UO_2 standard gave nearly identical results with the two fillings. However, owing to the L fluorescence radiation mentioned in subsection 2 d, this standard would be expected to give a higher counting rate with krypton than with argon, and these results may therefore signify that also the UO_2 counting rate was too high in run No. 1.

The increase in the photosensitivity of a G—M counter is, according to the results of H. NEUERT[37], proportional, within certain limits, to the number of discharges which have occurred in the counter during an interval of some tens of minutes before the instant considered. Since the photosensitivity of the counter decays rather quickly after a sudden decrease in the rate of the discharges, it may be expected to reach an equilibrium value during prolonged counting at a constant rate. This explains why the increase of counting rate due to the same amount of external light represents an approximately constant percentage of the counting rate in the measurements No. 1 of Tables 4 and 5.

In subsequent measurements it was checked that outside light was effectively excluded from the counter, by observing the fluctuations in the counting rate given by the Sr^{90} standard. No increase with counting time could be detected.

The absence of spurious pulses caused by possible faults in the electronic equipment was checked repeatedly during the measurements. This was done either by applying to the amplifier a high voltage exceeding the operating voltage of the counter, the latter being disconnected, or by reducing the counter voltage some tens of volts below the Geiger threshold. In the first case, no pulses were observed during intervals of the order of 24 hours. In the second case, about 0.02 and 0.05 counts/min. were counted with krypton, at 60 and 20 volts below the threshold voltage, respectively. With argon, the observed counting rates were only about half these values. The number of pulses below threshold was independent of the actual counting rate in the Geiger region. Other counters tested showed a similar effect.

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[35] K. H. L. LAUTERJUNG and H. NEUERT, Z. f. Phys. *122*, 266, 1944.

[36] M. CHAUDHRI and A. G. FENTON, Proc. Phys. Soc. London *60*, 183, 1948.

[37] H. NEUERT, Z. f. Naturforschung *3 a*, 221, 1948.

The most probable explanation is that these pulses are produced by α-particles from radioactive impurities present in the walls of the counter. Owing to the large ionization of the α-particles in the counter gas, the size of the α pulses is sufficient to give a count even though the gas amplification of the counter is relatively low for voltages below the Geiger threshold. Above this threshold, the number of α counts should, of course, be independent of the nature of the filling gas. The value deduced from these measurements for the number of α-particles emitted per hour per cm^2 of KF, KCl, or Al surface, is about 0.1 α/cm^2/hour. This value may be compared with the value of 0.07 α/cm^2/hour reported by HESS and VANCOUR for the brass walls of several ionization chambers.[38]

In order to detect the x-rays from A^{40}, measurements were made with the KF source with no Al foil on it, by alternating the argon and krypton fillings several times. The counting rate in each pair of measurements was consistently higher with krypton than with argon, the difference amounting always to about 0.4 counts/min. The alternate results obtained with argon filling are collected in row No. 3 of Table 4, the results of the alternate krypton measurements being given in row No. 4. The final difference of 0.42 ± 0.06 counts/min. found in these measurements would, according to the computed values given in Table 2 a, correspond to a branching ratio of about 0.5, which result is still about four times larger than the value of the γ/β ratio (0.127).[9]

The Sr^{90} standard, which was the only pure β source used in these measurements, yielded practically equal results with the two fillings (rows Nos. 3 and 4 of Table 5). With the UO_2 and thorite standards, differences of about 1 % and 5 % were found, respectively. As has been mentioned previously, these differences are caused by the soft x-rays or γ radiations emitted by these sources. Hence, they do not indicate different β-efficiencies of the counter with the two gases.

In an attempt to find the cause of the relatively large difference between the results obtained for KF with krypton and argon fillings, respectively, a rough calculation was made of the possible influence on the measurements of Bremsstrahlung[39], as well as of that of the continuous γ spectrum emitted by K^{40} during the process of β-decay (the so called "internal" Bremsstrahlung).[40, 41] The total intensity of these γ radiations is known to be small, but the proportion of low-energy quanta, effective in the present difference measurements, is relatively high. However, most of these quanta would reach the counter volume together with simultaneous β-rays, and consequently would give no difference in the counting rates yielded by the two filling gases.

For thin sources, such as those used in this work, calculation shows that the number of soft quanta in the continuous γ spectrum originating from the K^{40} nucleus exceeds by far that due to the deceleration of the β-particles of K^{40} in the Coulomb field of other atoms. Nevertheless, the difference to be expected as a result of this continuous nuclear γ spectrum could be shown to be less than 0.01 counts/min. in the conditions of the present experiments.

An estimate was also made of the influence of the characteristic x-rays pro-

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[38] V. F. HESS and R. P. VANCOUR, Phys. Rev. *76*, 1205, 1949.

[39] W. HEITLER, *The Quantum Theory of Radiation* (Oxford, 1944), second edition, p. 170.

[40] J. K. KNIPP and G. E. UHLENBECK, Physica *3*, 425, 1936.

[41] F. BLOCH, Phys. Rev. *50*, 272, 1936.

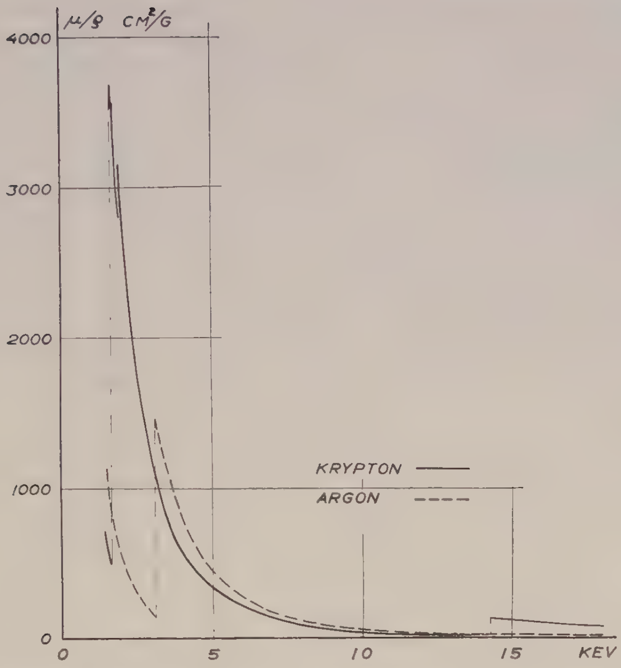

Fig. 7. Mass absorption coefficient of soft x-rays in krypton and in argon as a function of the quantum energy.

duced by the β-rays of K^{40} in other potassium atoms of the source. The probability of K shell ionization was computed by using the formula given by Mott and Massey.[42] This influence was found to be larger than that of the Bremsstrahlung spectrum, but the resulting counting rate difference should also in this case be less than about 0.01 counts/min.

It seemed possible to decide experimentally whether the counting rate difference observed with krypton and argon fillings could still be attributed to the K_α x-rays from potassium (3.31 KeV), or to a continuous γ spectrum, by using the method of sharp selective detection outlined in the following subsection.

4 b. Differential x-ray measurements of maximum selectivity

The selectivity of the differential counter method may be considerably improved by an appropriate choice of the krypton-argon pressure ratio.

The principle of the improvement is suggested by the curves of Fig. 7, which represent the variation of the calculated mass absorption coefficients of krypton and argon as a function of the quantum energy. These curves show that the linear absorption coefficients in the two gases can be made nearly identical over the whole energy region extending from the K absorption limit of argon (3.20 KeV) to that of krypton (14.3 keV). Consequently, between these two limits,

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[42] N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Oxford, 1949), second edition, p. 367.

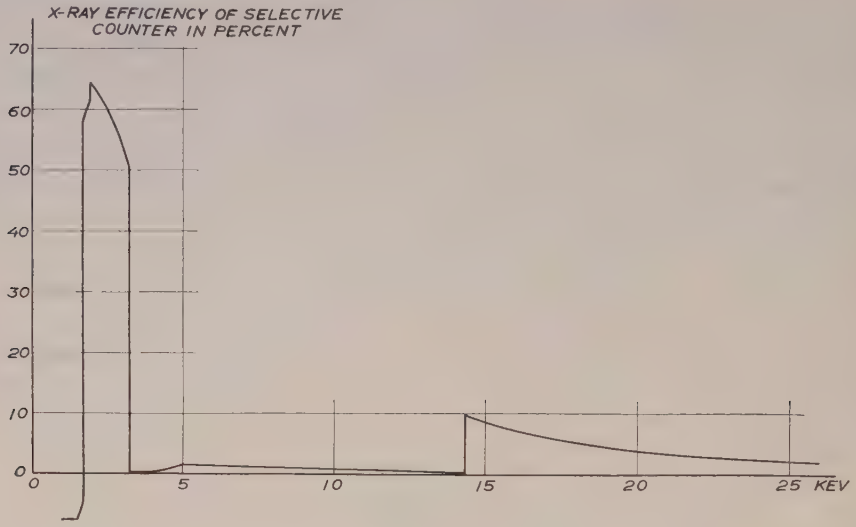

Fig. 8. Differential efficiency curve of the counter for x-rays. The selective properties of the counter are obtained by the use of compensated fillings.

the efficiency in difference measurements can be reduced to negligibly small values. At the same time, the differential efficiency in the region immediately below 3.20 keV and above 14.3 keV may be given almost any value up to unity, its value depending only on the absolute value of the krypton pressure and the distance traversed by the ray in the gas.

The curve of Fig. 8, calculated for a path length of 1.34 cm, an argon pressure of 185 mm and a krypton pressure of 128 mm, is given as an example for the differential efficiency obtainable between 1.5 and 25 keV. The above krypton-argon pressure ratio (0.69) was deduced from the ratio of the linear absorption coefficients in the two gases at N. T. P., immediately above the K absorption limit of argon. With this ratio, the counter efficiency below 1.68 keV (L_{III} absorption limit in krypton) turns out to be slightly greater in argon than in krypton. This explains the drop of the differential efficiency curve below the energy axis. However, no inconvenience results from this fact, for x-rays of energy less than 1.68 keV are greatly absorbed even by very thin source layers or counter windows.

This method of "compensated fillings" is in some respect complementary to the method of balanced filters of Ross[43], since it measures the absorbed intensities instead of the transmitted ones. A further difference between the two methods is that the absorbers used in the former are not adjacent in the periodic table. This makes it possible to compensate the counter efficiencies for a relatively large energy region.

The method may, of course, be extended to other pairs of fillings, usually rare gases.

Fairly accurate results may be expected for a radiation consisting mainly of x-rays. However, when the greatest part of the radiation consists of β-particles, as in the present experiments, even small variations in the β-efficiency of the counter must be taken into account.

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[43] P. A. Ross, J. Opt. Soc. Am. *16*, 433, 1928.

Measurements were made with the KF source at a krypton pressure of 128 mm of Hg in the counter. At this pressure the x-ray efficiency of the counter, for quantum energies between 3.20 and 14.3 keV, should be the same as with argon at 185 mm of Hg. The counting rate obtained, as indicated in row No. 5 of Table 4, is seen to be definitely lower than that observed with krypton at 185 mm (row No. 4), although the β-efficiency of the counter, checked by means of the external Sr^{90} standard, appeared to be practically the same (rows Nos. 4 and 5 in Table 5).

Table 6 gives in detail the results of this run, to illustrate the reliability of the counter measurements. As is seen, the standard error of the mean computed from the residuals of the individual measurements is practically equal to the standard deviation of the final result.

Table 6

Partial results with Kr at 128 mm of Hg. Run No. 5

Date	Counting time t in min.	Number of counts	Counts/min.
2/10	1 472	68 730	46.69 ± 0.18
3/10	1 384	65 173	47.09 ± 0.18
4/10	1 464	68 265	46.63 ± 0.18
5/10	1 480	68 981	46.61 ± 0.18
6/10	1 368	64 135	46.88 ± 0.19
7/10	1 568	73 557	46.91 ± 0.17
8/10	1 272	59 504	46.78 ± 0.19
9/10	1 408	65 794	46.73 ± 0.18
10/10	1 448	67 671	46.73 ± 0.18
11/10	1 472	68 845	46.77 ± 0.18
Total	14 336	670 655	46.78 ± 0.06

Standard error of the mean computed from the residuals x_i

$$\bar{\sigma}_m = \sqrt{\frac{\sum_{i=1}^{10} t_i \cdot x_i^2}{9 \sum_{i=1}^{10} t_i}} = 0.05 \text{ counts/min.}$$

The counting rates obtained with compensated fillings were nearly equal, as shown by rows Nos. 3 and 5 in Table 4. However, an unambiguous interpretation of this result requires the knowledge of the variation of the counter efficiency with pressure, in particular for the case of β-rays emitted from an internal source.

This question was therefore investigated both experimentally and theoretically. A brief account of the control experiments made is given in the next subsection. The calculated results and their comparison with experiment are presented in section 5.

4 c. Control measurements with KCl

Control measurements were made with the KCl source described in subsection 2 d. This source may be considered as a pure β emitter, since its γ radiation is negligible and the K_α x-rays from A^{40} are almost completely absorbed in the source itself.

The results obtained with krypton at three different pressures and with argon at 185 mm of Hg are listed in column 2 of Table 7. Two points may be noted concerning these results. The first is that the counting rate with krypton at a pressure of 285 mm was about 1.5 % higher than at 128 mm. The second, that the krypton-argon difference at 185 mm was 0.30 ± 0.14 counts/min., i. e., of the same order of magnitude as the difference observed with the KF source under similar conditions. Thus, the greater part of the latter difference must be due to some cause other than the argon x-rays.

Table 7

Measurements made with a KCl source of 4.12 mg/cm²

Rare gas pressure at 20 C in mm of H	Thickness of Al absorber		
	0 mg/cm² counts/min.	0.2 mg/cm² counts/min.	1.0 mg/cm² counts/min.
A 185	47.11±0.09	45.88±0.09	45.10±0.09
A 564	—	—	46.32±0.09
Kr 128	47.19±0.09	45.68±0.10	44.90±0.09
Kr 185	47.41±0.11	46.34±0.09	45.64±0.10
Kr 261	—	—	46.01±0.10
Kr 285	47.90±0.08	46.60±0.11	46.38±0.10
Kr 388	—	—	46.87±0.09

In order to verify that the radiation from the KCl source contains no x-rays of secondary origin, measurements were made with this source by placing thin Al foils (0.2 and 1.0 mg/cm²) between the source holders and the cathode windows. The results of these measurements are collected in columns 3 and 4 of Table 7.

The increase of counting rate with pressure is seen to be even more pronounced than without internal absorbers, especially with the thicker foil. The krypton-argon differences observed at 185 mm of Hg were, with these absorber foils also, of the same order of magnitude as for the KF source.

It is worth noting that the counting rate was higher with krypton at 388 mm than with argon at 564 mm of Hg, although this pressure ratio corresponds to compensation for x-rays between 3.20 and 14.3 keV, and the β-efficiency with krypton was certainly not higher than with argon, owing to the much lower krypton pressure.

The results obtained with the standard sources at the ends of the different runs of Table 7 are given in Table 8. In some of the measurements made with the Sr^{90} source an Al tube of 0.5 mm wall thickness was placed around the counter, for the purpose of reducing the high counting rate given by this source. As is seen, for the Sr^{90} standard at 185 mm of Hg about the same results

Table 8

Measurements made with the standard sources at the ends of the corresponding runs in Table 7

Rare gas pressure at 20° C in mm of Hg	Internal Al absorber mg/cm^2	Thorite 3 mm Al absorber γ counts/min	UO_2 No absorber β counts/min	Sr^{90} 0.5 mm Al absorber β counts/min	Sr^{90} No absorber β counts/min
A 185	0	751 ± 3	2019 ± 6	2988 ± 5	5376 ± 13
Kr 128	0	770 ± 3	2032 ± 6	2977 ± 7	5356 ± 13
Kr 285	0	813 ± 3	2070 ± 6	3021 ± 7	5432 ± 9
A 185	0.2	736 ± 3	2014 ± 8	2952 ± 6	5302 ± 13
Kr 128	0.2	748 ± 3	1991 ± 8	2953 ± 7	5313 ± 9
Kr 185	0.2	776 ± 4	2016 ± 8	2963 ± 7	5313 ± 13
Kr 285	0.2	793 ± 4	2032 ± 6	2974 ± 10	5331 ± 13
A 185	1.0	—	2004 ± 8	2975 ± 3	5366 ± 13
A 564	1.0	—	2076 ± 6	3049 ± 7	5440 ± 12
Kr 128	1.0	758 ± 3	2004 ± 8	2958 ± 7	5335 ± 10
Kr 185	1.0	782 ± 4	2026 ± 8	2976 ± 7	5339 ± 13
Kr 285	1.0	—	2045 ± 8	2996 ± 6	—
Kr 388	1.0	—	2060 ± 6	3019 ± 3	5365 ± 7

were obtained with krypton as with argon. With the UO_2 standard, for the same pressure, slightly higher counting rates were found with krypton than with argon, just as during the KF measurements.

The values in Table 8 are all a few percent higher than the corresponding values of Table 5, owing to the fact that the source holders for KCl were somewhat thinner than those used to support the KF sources.

The KCl measurements made with an Al foil of 0.2 mg/cm^2 are not quite comparable with the other measurements, for this very thin foil could not be placed tight enough over the whole surface of the source. It is probable that the counting volume was slightly reduced, owing to this cause, as is also suggested by the fact that the counting rates obtained were comparatively low even in the case of the γ standard (thorite).

A further discussion of the results of these control measurements will follow in section 5, after some theoretical considerations on the β-efficiency of G–M counters.

5. Efficiency of a GEIGER-MÜLLER counter for isotropically distributed electrons from a source deposited on the inside wall of the counter

5 a. Losses due to insufficient ionization

The efficiency of a G–M counter may become appreciably less than 100 % for electrons whose trajectories in the gas are too short, and which therefore may fail to produce a single ion pair along their path. Such comparatively low efficiency values were found previously by the author for monoenergetic electrons travelling nearly equal lengths in the counter.[44] In order to compute the counter efficiency in the conditions of the present experiments, in which the electron paths have different lengths, it is necessary to consider the relative probability of the different distances traversed by the electrons. This may readily be done if it is assumed that the angular distribution of the β-rays from a thin source layer of negligible absorption is isotropic.

The calculations will be restricted to the case of straight paths connecting the point of emission O on the cylindrical wall, with a point M on the same surface. If I denotes the average number of primary ionizing events along a distance equal to the diameter of the counter, and l the ratio of the distance OM to this diameter, the probability of no ionizing events occurring along a path OM, will be

$$e^{-Il}.$$

For simplicity, the cylinder will be assumed to have unit diameter so that $l = \text{OM}$.

Let $P(l)$ be the probability that the path length is $\leq l$. The loss of efficiency, i.e., the probability that an arbitrary electron will produce no ionization along its path, is then

$$q = \int_0^\infty e^{-Il} \cdot dP(l). \tag{16}$$

$P(l)$ may be expressed as the fraction of the hemisphere in which the path length is $\leq l$. Let θ denote the angle between OM and the normal, and α the dihedral angle shown in Fig. 6. From the relation

$$l = 1/(\cos\theta + \sin\theta \cdot \operatorname{tg}\theta \cdot \cos^2\alpha) \tag{17}$$

it follows that

$$\alpha = \arcsin\sqrt{\frac{1-(1/l)\cos\theta}{1-\cos^2\theta}}. \tag{18}$$

For reasons of symmetry it is sufficient to consider the first quadrant, $\frac{\pi}{2}\sin\theta \cdot d\theta$, of the elementary solid angle $2\pi\sin\theta \cdot d\theta$. The solid angle containing only paths $\leq l$ in this quadrant is $\alpha \cdot \sin\theta \cdot d\theta$, so that by replacing α by its value from (18), one obtains

$$P(l) = \frac{2}{\pi}\int_0^l \arcsin\sqrt{\frac{1-(1/l)\cos\theta}{1-\cos^2\theta}} \cdot d(\cos\theta). \tag{19}$$

This function has been computed by numerical integration of (19). It is represented by the curve of Fig. 9. For a counter of diameter D, the symbol l should, of course, be interpreted as the ratio of the distance OM to D.

Within limited intervals, in which dP may be considered as proportional to dl, $dP = a_i \cdot dl$, and integration of (16) yields

$$q_i = \frac{a_i}{I}(e^{-Il_{i-1}} - e^{-Il_i}). \tag{20}$$

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[44] T. GRÁF, J. Phys. Radium *10*, 513, 1939.

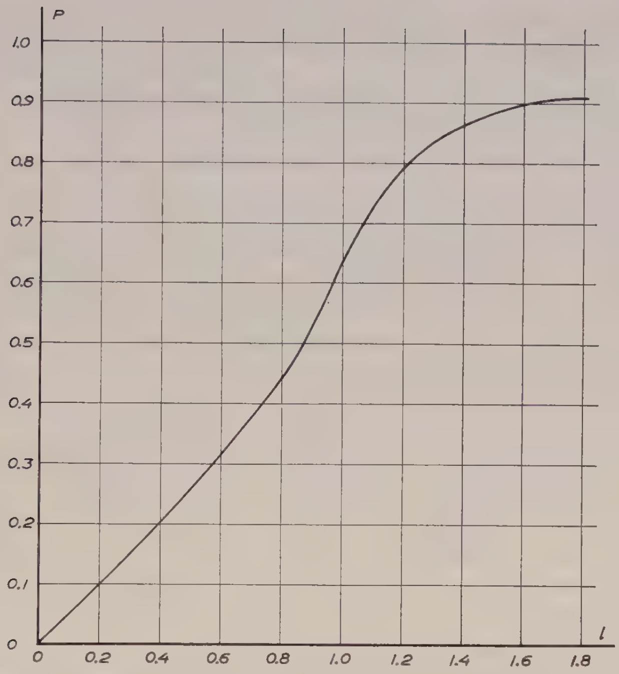

Fig. 9. Probability of a ratio $OM/D \leq l$ for a right circular cylinder. OM = distance between two points on the cylindrical surface. D = diameter of the cylinder.

Table 9

i	Interval Δl	a_i	q_i
1	0 —0.1	0.500	0.0302
2	0.1—0.2	0.505	0.0102
3	0.2—0.3	0.51	0.0035
4	0.3—0.4	0.52	0.0012
5	0.4—0.5	0.54	0.0004
6	0.5—0.8	0.61	0.0002
		$\sum_{i=1}^{6} q_i$ =	0.0457

Table 9 gives, as an example, the contribution to q of the different intervals of l, for $I = 11.0$. This value of I corresponds to a counter of 1.34 cm inner diameter filled with argon and alcohol to 185 and 15 mm of Hg, respectively. It would seem reasonable to assume that the β-efficiency of this counter is very nearly 100 %. It is therefore rather surprising to find that for an isotropic distribution the computed loss of efficiency amounts to as much as 4.6 %.

The expression

$$\varepsilon = 1 - \sum_{i=1}^{n} q_i, \tag{21}$$

i.e., the probability that an arbitrary electron emitted by a internal source will produce at least one ion pair in the filling gas, was computed for different values of I. It is represented by the solid curve in Fig. 10. This curve gives thus the calculated value of the β-efficiency for any value of the counter diameter or the gas pressure within the range of I represented.

5 b. Influence of other factors

When comparing the calculated values of the β-efficiency with the experimental results, one must bear in mind that, besides insufficient ionization, there are a number of other causes which may influence the efficiency losses.

As has been shown by D. H. WILKINSON[45], the probability that a single ion pair will initiate a discharge in the counter may be less than unity. This reason may also explain the fact that the values of the primary specific ionization in air, deduced from earlier efficiency measurements of the author, were 20—30 % lower than the theoretical values. These measurement were made with two G—M counters in coincidence, mounted inside a β-spectrograph.[44]

A further loss of efficiency may be expected when the source, or some part of it, is placed too near to the insensitive end region of the counter. In the light of the results of Table 9 the efficiency loss due to end effects should be negligible if the source is at a distance larger than 0.5 D from the ends of the effective counter volume.

Whereas the causes just mentioned tend to increase the efficiency losses, other factors, such as absorption in the source, scattering in the gas, or back-scattering from the counter walls, will act in the opposite direction. Scattering and back-scattering will reduce the losses of efficiency by lengthening the electron trajectories inside the gas volume. Absorption will cause a reduction in the relative abundance of the short paths.

5 c. Primary specific ionization

The primary specific ionization of β-rays, denoted by I_p, reaches a minimum value for an electron energy of about 2 MeV. The variation of I_p around the minimum is, however, so slow, that for the present purpose I_p may be considered constant over a large energy region, from about 0.7 MeV upwards. In order to account for the larger ionizing power of the low-energy β-rays of the spectrum it seems advisable to adopt for I_p a value slightly larger than its minimum value.

One may take the values of I_p determined by S. C. CURRAN and J. REID for 0.42 MeV electrons in argon and in ethyl alcohol.[46] These values, namely, 29.8 in argon and 79.0 in ethyl alcohol, at N. T. P., are, in point of fact, the only ones available in this energy region. They were obtained by the use of the previously mentioned coincidence method.[44]

For krypton no determinations of I_p seem to have been made at all. An approximate value for krypton may be deduced from the value of I_p for argon, by assuming that in the two gases I_p is inversely proportional to the mean energy, W, necessary for producing one ion pair. This relation was found to hold reasonably well for argon and air, for example. With $W_A = 28.2$ eV, and

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[45] D. H. WILKINSON, Phys. Rev. *74*, 1417, 1948.

[46] S. C. CURRAN, and J. REID, Nature *160*, 866, 1947.

W_{Kr} 26.2 eV, deduced from measurements on the ionization of α-rays[47], the primary specific ionization in krypton for 0.42-MeV electrons is found to be 32.1 primary ion pairs/cm at N. T. P.

5 d. Comparison with experiment

In order to make a comparison possible between the calculated values of the β-efficiency and the experimental results listed in Table 7, the differences between the counting rates obtained at different pressures were expressed as percentages of the number of true β counts. This latter was estimated at 40 β/min., in round numbers. The experimental differences obtained in this way are shown in Table 10, together with the computed β-efficiency values and their differences.

Table 10

Rare gas pressure in mm of Hg at 20° C	I at 20° C	Computed β-efficiency in %	Computed differences in %	Exp. differences in %		
				1.0 mg/cm^2	0.2 mg/cm^2	0 mg/cm^2
A 185	11.0	95.43	0	0	—	—
A 564	29.5	98.23	2.80	3.05	—	—
Kr 128	8.7	94.20	0	0	0	0
Kr 185	11.7	95.72	1.52	1.85	1.65	0.55
Kr 261	15.7	96.80	2.60	2.77	—	—
Kr 285	17.0	97.04	2.84	3.70	2.30	1.78
Kr 388	22.4	97.79	3.59	4.93	—	—

The quantity I in column 2 represents the average number of primary ionizing events along a distance of 1.34 cm, equal to the diameter of the counter. It was computed for the rare gas pressures indicated in column 1, and an additional alcohol pressure of 15 mm of Hg, by using for I_p the values given in the previous subsection.

In Table 10 the counting rates obtained at the lowest argon and krypton pressures were taken as zero points for the experimental differences. Similarly, the experimental curves, representing these differences in Fig 10, were normalized at the points of the computed curve which correspond to a pressure of 128 mm of Hg for krypton ($I = 8.7$), and 185 mm for argon ($I = 11.0$). Apart from this latter point of normalization, there is only one further experimental point for argon, at $I = 29.5$, corresponding to a pressure of 564 mm. Since this point is very close to the computed curve, it seems reasonable to draw the argon curve through the two experimental points in the manner indicated in the figure.

It is apparent from a comparison of the experimental krypton and argon curves that the increase of the krypton curve obtained with 1.0 mg/cm^2 Al is far too steep, and that this increase results in excessive values of the β-efficiency. On the contrary, although insufficiently defined, the argon curve seems to be compatible with an asymptotic efficiency value of 100 %

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[47] G. Stetter, Z. f. Phys. *120*, 639, 1943.

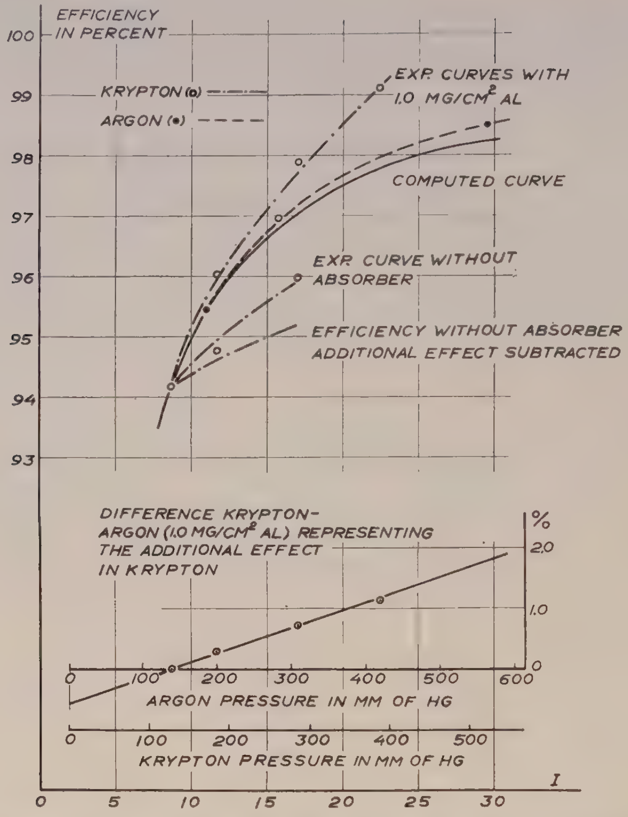

Fig. 10. Computed counter efficiency for the β-rays of an internal source as a function of the average number of primary ion pairs, I, produced along the diameter of the counter (full curve), together with the observed increase of the counting rate as a percentage of the total number of β-rays (40/min.). The experimental curves are fitted to the computed curve at $I = 8.7$ for krypton (128 mm of Hg), and at $I = 11.0$ for argon (185 mm of Hg). The lower curve represents the "additional effect" in krypton as a function of I.

A thorough consideration of the different factors influencing the counter efficiency makes it clear that the variation of the β-efficiency as a function of I must be very nearly the same for the two gases. Consequently, the steeper increase observed with krypton has to be attributed to some hitherto not considered "additional effect". A measure of this "additional effect" is given by the difference of the krypton and argon curves. As may be seen in the lower part of Fig. 10, the differences formed for different values of I define approximately a straight line.

The krypton curve obtained without internal absorber is defined by three points only. Its slope is smaller than that of the krypton curve obtained with 1.0 mg/cm² Al, even though the actual counting rates do not appear to imply that the values of the β-efficiency at the higher pressures are too low. It is possible that owing to a greater back-scattering from KCl than from Al the whole curve is displaced towards higher efficiency values, for which it would be expected to find a smaller slope. Since part of the increase shown by this krypton curve is possibly due to the same "additional effect" as was found with 1.0 mg/cm^2 Al on the source, the ordinates of the straight line representing the "additional effect" were subtracted from the corresponding ordinates of the experimental krypton curve. The difference, which is represented by the lowest broken curve in Fig. 10, gives then the variation of the β-efficiency alone. This variation is seen to be about equal to that observed with the external Sr^{90} standard (Table 8). It is, however, smaller than the increase of the computed curve, or than the experimental increase found with 1.0 mg/cm^2 Al in the case of argon filling.

Approximate values of the corrections due to the variation of the β-efficiency in the KCl measurements are given in Table 11 as a function of the value of I at different gas pressures. They were deduced from the lowest curve in the upper part of Fig. 10 by assuming the same number for the true β counts as was assumed earlier in the inverse transformation.

Table 11

Increase of counting rate due to the variation of the β-efficiency, relative to its value at $I = 8.7$, and the "additional effect" in Kr relative to A, as a function of I

Rare gas		Kr	A	Kr	Kr
Rare gas pressure in mm of Hg		128	185	185	388
I, primary ion pairs along the counter diameter		8.7	11.0	11.7	22.4
β-efficiency correction in counts/min	KCl, no absorber	0	0.13	0.17	0.54
	KF, 0.7 mg/cm^2 graphite	0	0.18	0.24	—
"Additional effect" in counts/min		0.23	—	0.34	0.70

The "additional effect", expressed in counts/min., was deduced from the straight line of Fig. 10 in a similar way. Its value at 128 mm of Hg was found to be 0.23 counts/min. by extrapolating the straight line to zero pressure. This amount was added to the values deduced at other pressures, which, in reality, only represent the increase of the "additional effect" relative to its value at 128 mm of Hg.

For the KF source, the β-efficiency correction was deduced from the experimental results Nos. 4 and 5 of Table 4, which indicate a difference of 0.35 ± 0.08 counts/min. between the measurements made with krypton at 128 and 185 mm of Hg. Part of this difference is, however, due to the variation of the "additional effect" for the two pressures, amounting, according to the figures in the last row of table 11, to about 0.11 counts/min. The error in this latter value is estimated to be about 30 %. The β-efficiency correction between the KF measurements made at these two pressures is thus 0.24 ± 0.09 counts/min. From this value the corresponding correction for argon at 185 mm ($I = 11.0$) relative to krypton at 128 mm of Hg ($I = 8.7$) was deduced by assuming a linear variation of the correction with I.

In conclusion, the preceding considerations show that for the measurements made with KF at identical krypton and argon pressures, the correction due to the variation of the β-efficiency will not exceed about 0.06 counts/min. The greater part of the difference of 0.42 ± 0.06 counts/min., obtained in these measurements, has to be attributed to an "additional effect" which the counter exhibits only when it is filled with krypton.

In the difference measurements made with krypton at 128 mm and argon at 185 mm of Hg (compensated fillings), the β-efficiency correction amounts to about 0.18 counts/min. However, the relative loss of β-efficiency with krypton at 128 mm of Hg is approximately cancelled by an "additional effect" of about 0.23 counts/min.

The differences which remain after taking into account the variation of the β-efficiency and the "additional effect" are for both pairs of measurements very small, of the order of 0.02 counts/min. The intensity of the argon x-rays, to which these corrected differences may be attributed, has therefore an upper limit given only by the statistical counting errors combined with the uncertainties in the above corrections.

6. Results

6 a. Explanation of the "additional effect" in krypton-filled counters

Two kinds of causes were considered as possible explanations of the "additional effect": (1) x-rays of quantum energy between 14.3 and, say, 30 keV, originating either inside the counter or in the lead shielding, and (2) spurious counts which occur only when the counter is filled with krypton.

Table 12

γ-ray measurements. Scale-of-two. Thin-walled counter (0.01 cm of Al) of 4.0 cm length and 1.6 cm I.D. Absorber cylinder of Perspex, 1.8 cm I.D. and 2.5 cm O.D. Source cylinders of 5.0 cm length, 2.6 cm I.D. and 3.4 cm O.D.

Sources	Net rates in counts/min. (background subtracted)	
	A 565 mm of Hg	Kr 385 mm of Hg
KCl	6.3 ± 0.2	7.9 ± 0.3
$KBrO_3$	3.5 ± 0.2	4.2 ± 0.3
KJ	3.8 ± 0.2	5.3 ± 0.2

In order to investigate (1), special experiments were carried out with a conventional G–M counter having a wall thickness of 0.01 cm of Al. As sources, hollow cylinders of pressed potassium salts were used, which weighed between 30 and 50 grams.[48] The β-rays from these sources were absorbed by a Perspex tube of 0.35 cm wall thickness, placed concentrically between the counter and the source. Any soft x-rays should be detected by the alternate use of krypton and argon in the counter. The net counting rates obtained, due mainly to the 1.46 MeV γ radiation of K^{40}, are shown in Table 12. The differences found between the krypton and argon measurements are too large to be due to a variation of the counter efficiency for this hard γ radiation. They can, however, be explained by Bremsstrahlung produced in the source cylinders by the β-rays of K^{40}. From these measurements it was deduced that the number of quanta of energy greater than 14.3 keV, due to Bremsstrahlung, or to any other cause, must be less than 0.01 per decaying K^{40} atom. It follows that the number of such quanta emitted from the thin internal source layers used in the main experiments must be far too small to account for the "additional effect" observed.

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[48] E. GLEDITSCH and T. GRÁF, Archiv f. Math. og Naturvidenskab *44*, Nr. 14, 145, 1941.

In another experiment, the background of the same Al counter was measured with and without a brass cylinder of 0.4 cm wall thickness, placed around the counter, inside the lead shielding. This cylinder would absorb any soft x-rays which might be emitted from the lead. The β-rays from radioactive impurities in the lead (3–4 β/min.) were, as in all previous experiments, absorbed by a bakelite cylinder of 0.25 cm wall thickness. The results of these background measurements, given in Table 13, show that no measurable soft radiation is emitted from the lead.

Table 13

Background measurements. Scale-of-two. Same counter as in Table 12. Brass cylinder of 0.4 cm of wall thickness

	Counts/min. A 565 mm of Hg	Counts/min. Kr 385 mm of Hg
Without brass cylinder	4.65 ± 0.07	5.12 ± 0.06
With brass cylinder	4.66 ± 0.15	5.03 ± 0.09

The same measurements show, however, that the background rates were in both cases about 10 % higher for krypton than for argon. In the light of the measurements of Table 3 this result appeared to be rather puzzling. A new determination was therefore made of the background rate of the internal-source counter, first at relatively high pressures, and then at 185 mm of Hg, the pressure used in the main measurements. The results, contained in Table 14, show that the background rate with krypton at 385 mm was about 10 % higher than with argon at 415 mm of Hg, the argon rate being practically the same as its value at 185 mm of Hg, obtained in the earlier measurements (Table 3).

The difference of 10 % agrees with that found with the thin-walled Al counter, but seems to contradict the results listed in Table 3. The reason for this ap-

Table 14

Background measurements made with the counter of Fig. 3

No.	Scale	Rare gas pressure in mm of Hg at 20° C	I at 20° C	Counts/min.	Differences in counts/min.
1	2	A 415	22.3	8.42 ± 0.05	0.85 ± 0.09
2	2	Kr 385	22.3	9.27 ± 0.07	
3	2	A 185	11.0	8.40 ± 0.05	0.29 ± 0.06
4	2	Kr 185	11.7	8.69 ± 0.04	
5	1	Kr 185	11.7	8.47 ± 0.02	

parent discrepancy was found to lie in the fact that during the early background measurements no scaling circuit was used, whereas the new determinations were made with a scale-of-two. This was confirmed by subsequent background measurements with krypton at 185 mm of Hg, carried out alternately with and without a scaling circuit. The difference between these measurements, also indicated in Table 14, is too large to be due to counting losses in the mechanical register, and can only be explained by the occurrence of an unexpectedly large percentage of double pulses. It is even more surprising that the values of Table 11 deduced for the "additional effect" at 185 and 388 mm of Hg, are found to be nearly equal to the corresponding differences between the background rates listed in Table 14. This agreement proves that assumption (2) is the right one, but shows, at the same time, that the double pulses occurring in krypton are of a special kind. Their number does not increase with the counting rate, in contrast to the spurious pulses usually encountered in G–M counters. This feature explains also why no spurious pulses exceeding in number 0.1 % of the counting rate of the standard were found with the anticoincidence method described in subsection 2 c.

Such double background pulses do not seem to have been noted in previous work with krypton-filled x-ray counters.[49, 50]

An investigation of the physical cause of these double pulses is outside the scope of this work. It may, however, be pointed out that they cannot be due to the special construction of the counter used, since the same relative increase of the background rate was found even for a counter of conventional design when it was filled with krypton instead of argon.

6 b. Concluding remarks on the krypton-argon difference method

As a result of this investigation, it has been established that a G–M counter filled alternately with krypton and argon is a suitable instrument for the quantitative and selective determination of extremely weak x-ray intensities (exept in the energy region between the K absorption limits of the two gases), provided that attention is paid to the following points.

(1) The counter should be sufficiently shielded in order to minimize the influence of external γ radiations.

(2) The same overvoltage (of about 60 volts) should be applied to the counter for both gases.

(3) The pressures for the two gases should be chosen to secure equal values of the primary specific ionization for β-rays (in the case of a large β-ray background), or equal values of the x-ray absorption coefficients between the two K absorption limits (in the case of a background due mainly to x-rays of quantum energy between 3.2 and 14.3 keV).

(4) When the effects to be measured are small compared with the background rates, these rates should be subtracted from the results, or what is still better, differences in the background rates should be eliminated by means of a quenching circuit suppressing counts due to double pulses.[51] It is not impossible that

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[49] G. Brogren, Ark. f. Mat., Astr. o. Fysik, *36 A*, No. 18, 1949.

[50] H. Friedman, Proc. I. R. E. *37*, 791, 1949.

[51] E. H. Cooke-Yarborough and C. D. Florida, J. Sci. Inst. *26*, 124, 1949.

the double pulses observed with krypton could also be suppressed by the use of another quenching vapour instead of ethyl alcohol.

As has been shown in this work, the counting rates, in the absence of soft x-rays, may be reproduced to within about 0.2 %. This accuracy is rather remarkable in view of the fact that the filling gases used in the alternate measurements are different and involve the use of different counter voltages. The ultimate sensitivity of the method for x-rays, the intensity of which is given as a difference between two measurements, is only limited by the above uncertainty.

Extremely small x-ray intensities may also be measured with a proportional counter[31], or a single x-ray counter combined with a magnetic field for the purpose of eliminating the β-ray background. For some applications the present method may prove to be more advantageous, on account of its greater stability and simplicity.

6 c. Electron capture/β-decay ratio in K^{40}

The intensity of the K_α x-rays from argon may be deduced, in the most direct manner, from the difference between the results yielded by the krypton-argon measurements for KF and KCl. These differential measurements were made at practically the same rate, at the same pressure, and with the same scaling circuit (scale-of-eight). A difference between their results cannot therefore be due to differences in the β-efficiency or in the background rate of the counter, and must be attributed to the difference in the number of the x-ray quanta emerging from the two sources. In fact, calculation shows that about 40 % of the K_α x-rays emerge from the KF source, as against 13 % from KCl.

Krypton-argon measurements made at a pressure of 185 mm of Hg yielded, as a result for the KF source, a difference of 0.42 ± 0.06 counts/min. (Table 4). For KCl, under the same conditions, 0.30 ± 0.14 counts/min. were obtained (Table 7, 0 mg/cm^2 Al). With the above interpretation and according to the computed data in Tables 2 a and 2 b, the difference between these two results would correspond to a branching ratio of 0.2 ± 0.3.

A more accurate value of the branching ratio may be obtained by subtracting the β-efficiency correction and the background correction from the result furnished by the KF measurements alone. As may be seen from Table 11, the β-efficiency correction between argon and krypton at 185 mm of Hg amounts to about 0.06 counts/min. The error in this correction is estimated to be about 50 %. The difference between the background rates obtained with argon and krypton, at the same pressure, was found to be 0.29 ± 0.06 counts/min. (Table 14).

With these corrections, the difference of 0.42 ± 0.06 counts/min. found for the KF source is reduced to 0.07 ± 0.09 counts/min. According to the calculated relations in Table 2 a, this corrected difference corresponds to a branching ratio of 0.08 ± 0.11.

The latter value, i.e., an upper limit of about 0.2 for the branching ratio, is to be considered as the final result of the present investigation.

This result does not take into account the uncertainty in the value of the fluorescence yield (0.12). It is still sufficiently accurate to preclude with certainty the high values found previously by some authors[1-3], and to show, in agreement with the assumption first advanced by SUESS[5], that the number of capture processes does not differ appreciably from the number of γ quanta emitted by K^{40}.

Quite recently, after the present investigation was practically completed, several papers were published concerning the branching ratio in K^{40}.

One of these papers, due to SAWYER and WIEDENBECK[33], describes experiments in which extremely thin source layers containing potassium enriched in isotope 40 were used. In these experiments it was possible to detect the Auger electrons emitted following the capture process. From the number of Auger electrons a value of 0.135 was deduced for the branching ratio, the estimated error in the result being about 30 %.

Even though this result should be increased to about 0.146, to account for the L_I-capture process, it is still in good agreement with the value of the γ/β ratio (0.127).[9] A similar measurement is, as stated in subsection 2 a, not feasible with natural potassium.

Measurements were made by HOUTERMANS, HAXEL and HEINTZE[34] with two semi-cylindrical counters separated from each other by an Al foil which supported the K^{40} source on one of its sides. From a study of the difference between the counting rates in the two counters as a function of the gas pressure, these authors were able to deduce an upper limit of about 0.55 for the branching ratio. They noticed that when the counters were filled with krypton the difference between the counting rates on the two sides of the Al foil continued to increase even for the highest pressures used, but could give no explanation of this effect.

CECCARELLI et al.[52] used a counter placed in a strong magnetic field to prevent the electrons from reaching the counting volume. Inserting a 0.03 mm thick Al foil between the source and the wires forming the cathode of the counter, these authors observed a decrease of 0.15 ± 0.08 counts/min. for a background rate of about 34 counts/min. When attributed to the K_α x-rays from argon, this difference corresponds, according to the authors, to a branching ratio having an upper limit of 0.07. The relatively low value of the above difference may, in reality, be due to the fact that the counting rate with Al shield included additional α counts which were not eliminated by the magnetic field. This effect of the radioactive impurities of Al, discussed in subsection 4 a, may, in the experiments of these authors, correspond to as much as 0.20 counts/min., for example.

Recent simultaneous A^{40} and Ca^{40} determinations in 10^8-year old sylvite by INGHRAM et al.[53], by means of a mass-spectrometric isotopic dilution method, yielded a value of 0.126 ± 0.005 for the branching ratio. This value, resulting from a direct determination of the decay products of K^{40}, must be considered as the best value available at present.

It is also in excellent agreement with the value of 0.127 ± 0.012 obtained by the author for the γ/β ratio.[9] The latter ratio was deduced from the specific γ activity, for which the value of 3.4 ± 0.3 quanta per gram potassium per second was found*, and from the value of the specific β activity, 26.8 ± 1.2 β/g/second, already discussed in subsection 3 b.

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[52] CECCARELLI, QUARENI, and ROSTAGNI, Phys. Rev. *80*, 909, 1950.

[53] INGHRAM, BROWN, PATTERSON, and HESS, Phys. Rev. *80*, 916, 1950.

* In reference 21 this value was erroneously given as 3.4 ± 0.5.

This value of the γ/β ratio has been confirmed by the more recent determinations of SAWYER and WIEDENBECK[33] and of HOUTERMANS el al.[34]

With the above values of the specific activities, the total half-life of K^{40} is found to be $(1.33 \pm 0.08) \cdot 10^9$ years. When this value is used in the calculation of the rate of radioactive heat production, the rates during the earliest geological times turn out to be much smaller than their previously computed values.[8, 9] Even so, the heat generated in the earth's crust by potassium, say, 3.10^9 years ago, was of about the same importance as the amount of heat produced there by the radioactivity of uranium or thorium.

SUMMARY

1. An internal-source G—M counter has been constructed for the purpose of measuring the branching ratio between electron capture and β-decay in K^{40}. The x-rays resulting from the capture process should be detected by differential measurements in which the counter is filled alternately with krypton and argon.
2. In order to make possible the measurement of counting rate differences of the order of 0.2 %, methods have been evolved for improving and checking the reliability of the counter operation.
3. A method of computing the number of x-ray counts is outlined, and a simple approximate formula is given for the x-ray efficiency of internal-source counters.
4. It is shown that optimum selectivity may be secured for x-ray measurements by the choice of an appropriate krypton-argon pressure ratio. The differential efficiency of the counter is then practically zero in the whole energy region between 3.2 and 14.3 keV. It is high between 1.7 and 3.2 keV, and in the region immediately above 14.3 keV.
5. The value of the branching ratio may be deduced, without the necessity of corrections, from the results obtained under identical conditions with KF and KCl, in which sources the x-rays are absorbed to different extents.
6. Measurements made with KCl at different pressures show, that the background rates with krypton and argon may be made equal by using a pressure ratio corresponding to equal values of the primary specific ionization for β-rays in the two gases, and by suppressing the slight increase in the krypton background rate, found to be due to double pulses of a special kind.
7. A calculation of the β-efficiency of internal-source counters has been made, and the results compared with those furnished by the experiments.
8. The results obtained with KF yield, when corrected, an upper limit of about 0.2 for the branching ratio. This upper limit is compatible with an exact value equal to that of the γ/β ratio, given by an earlier determination of the author as 0.127 ± 0.012. With this value, and with the previously measured value of the specific β activity of potassium, the total half-life of K^{40} is found to be $(1.33 \pm 0.08) \cdot 10^9$ years.

The author wishes to thank Professor L. MEITNER for her continued interest in this work and for many critical discussions.

His thanks are also due to Mr C. TAYLOR for discussions and for linguistic advice concerning the manuscript.

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Tryckt den 23 april 1951

Uppsala 1951. Almqvist & Wiksells Boktryckeri AB